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14. ABSTRACT  A multi-prong program of study has been conducted that encompassed nonlinear optical material synthesis, quantitative characterization, design and proof of concept demonstration of practical devices for eye/sensor protection against agile frequency lasers in the entire visible spectrum. Significant breakthroughs have been achieved in developing supra-nonlinear liquid crystalline films that possess extraordinarily large photorefractive responses, low switching thresholds and useful properties not available in other existing photorefractive materials. A class of multifunctional nonlinear organic liquids that are optically transparent in the entire visible spectrum for low level light, but possess large and very broadband multi-photon absorption and excited-state-absorption coefficients has been developed. These properties enable the construction of image transmission fiber arrays that could function as optical limiters against visible picoseconds – nanoseconds lasers with low operation power threshold and large dynamic operation range. Paralleling these material and devices studies, complete quantitative models for the molecular photonics, nonlinear laser propagation and device operational principles have been developed. The models accounted for all the nonlinear multi-photon absorption processes, as well as nonlinear scattering and concomitant thermal/density effects that are useful for longer time scales limiting applications, and enable identification of the optimal material composition and design of high performance all-optical devices.				
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## 1. Executive Summary

A multi-prong program of study has been conducted that encompassed nonlinear optical material synthesis, quantitative characterization, design and proof of concept demonstration of practical devices for novel optical filters, switches, image processing, and eye/sensor protection against agile frequency lasers in the entire visible spectrum. Specific projects performed in the program are broadly classified as follow: (i) Synthesis of nonlinear nematic liquid crystals containing highly photosensitive and/or photoconducting nano-particulates (Gold nano-wires, CdSe nanorods) and observation of several nonlinear optical processes potentially useful for image processing and optical limiting applications; (ii) Fabrication of nonlinear liquid crystal filled 2- and 3-D photonic crystals and polymer dispersed liquid crystalline photonic crystals and demonstration of widely tunable reflection spectra; (iii) Investigation and characterization of the mechanisms responsible for the supra- nonlinearities of these liquid crystalline systems and identification of the roles of various fields, molecular and optical parameters for optimum nonlinearity, response time and other performance characteristics; (iv) Synthesis of neat nonlinear liquids with large two- and multi-photon absorption and excited-state absorption coefficients and characterizations of these liquids; (v) Demonstration of optical limiting capabilities of the organic liquids against picosecond and nanosecond laser pulses; (vi) Fabrication of nonlinear fiber arrays with these nonlinear liquids as wave-guiding cores and evaluation of the image transmission characteristics of the fiber arrays; (vii) Quantitative modeling of self-action effects such as all-optical switching, optical limiting, nonlinear transmission of (fiber)guided visible as well as infrared lasers.

The results and findings obtained in these projects culminated in 2 Ph. D. theses, 19 refereed journal papers, 21 technical conference presentations/proceedings, 1 invention disclosure and 1 book. Significant breakthroughs have been achieved in developing supra-nonlinear liquid crystalline films that possess extraordinarily large photorefractive responses, low switching thresholds and useful properties not available in other existing photorefractive materials. A multifunctional neat nonlinear organic liquid and its derivatives that are optically transparent in the entire visible spectrum for low level light, but possess large and broadband multi-photon absorption and excited-state-absorption coefficients have been developed. These properties enable the construction of image transmission fiber arrays that could function as optical limiters against visible picoseconds – nanoseconds lasers with low operation power threshold and large dynamic operation range. Paralleling these material and devices studies, complete quantitative models for the molecular photonics, nonlinear laser propagation and device operational principles have been developed. The models accounted for all the nonlinear absorption processes such as Reverse Saturable Absorptions [RSA], Two-Photon Absorption [TPA] and Excited State Absorption [ESA], as well as nonlinear scattering and concomitant thermal/density effects that are useful for longer time scales limiting application. In conjunction with quantitative analysis of the experimental results, these models enable the identification of the optimal material composition and the design of high performance next generation optical limiter and all-optical switches. The findings from this program point to promising future research and development possibilities on multiple time scale agile frequency switches and image/beam processing materials and devices.

## 2. Technical accomplishments- Narrative

### 2.1 Optical Nonlinearities of Liquid Crystals and Neat Organic Liquids - Overview

Advances in optical science and technology are enabled mainly by materials with unique functionalities in conjunction with specialized optical structures and processes. In the context of nonlinear optics, liquid crystals occupy an important niche as a result of their unique properties [1]. Besides their broadband birefringence and transparency, abilities to self-assemble into various crystalline phases and to conform to various flexible forms and shapes [e.g. photonic crystals, holey fibers, and nanostructures], liquid crystals are compatible with almost all other optoelectronic materials and technology platforms [1-4]. They are also laser-hardened. Impurities-free liquid crystals are capable of handling very intense pulsed lasers or high power continuous wave lasers [5, 6]; studies have shown that common liquid crystals such as 5CB and E7 can withstand nanosecond laser pulse of  $\sim 10$  Joules/cm<sup>2</sup> (corresponding to an intensity  $\sim 10^{10}$ W/cm<sup>2</sup>), thus making them particularly useful for constructing high power laser optics such as polarization rotators, wave plates, optical isolators and laser blocking notch filters and the optical limiter and sensor protection devices we intend to develop in this proposed program. Under extended illumination of the liquid crystals [E7 or 5CB] with cw laser intensity of several kilowatt/cm<sup>2</sup>, e.g. in stimulated orientation scatterings with focused lasers, liquid crystals also do not suffer any structural/chemical damages [6].

In their isotropic (liquid) phase [7, 8], liquid crystals also possess extraordinarily large electronic optical nonlinearities. In our program, we have developed an entire class of neat liquids (isotropic liquid crystals) [7] that possess extraordinarily large and ultrafast [response time < picoseconds] optical nonlinearities arising from a unique and almost ideal intra-molecular multi-photonic transition structures. In nematic liquid crystals, laser induced director axis reorientation and order parameter changes give rise to nonlinear refractive index coefficients that are orders of magnitude larger than all known nonlinear optical materials [1, 9-11]. Such collective optical nonlinearities are characterized by response times that can vary from *nanoseconds to milliseconds or longer*, depending on the degree of order [1]. In most cases, the nonlinearities are not dependent on the optical wavelength/frequency, thus enabling their applications over a very broad (from visible to infrared) spectrum in which nematic liquid crystal exhibits large birefringence. Table 1 compares and contrasts the optical nonlinearities of the isotropic and nematic phases of liquid crystals to other existing optical materials, and illustrates some of their unique and useful properties.

In the course of our program, we have made important breakthroughs and discoveries in both mesophases of liquid crystals. Details of some exemplary accomplishments are given in the following sections in accordance with the time frame in which they are performed. Specifically, in the initial phases of the program, most attention were paid to investigating the photorefractivity of nematic liquid crystals doped with photosensitive nano-dopants, while in the later phases of the program, studies are centered around nonlinear absorption processes in a isotropic-phase liquid crystal (neat liquid) and its use as fiber cores for broadband and multiple time scale optical limiting applications. Studies of the optical limiting and laser hardening applications of nematic liquid crystals have been conducted throughout the program.

Specific projects that have been successfully conducted are broadly classified as follow: (i) *Synthesis of nonlinear nematic liquid crystals containing highly photosensitive*

and/or photo-conducting nano-particulates (Gold nano-wires, CdSe nanorods) and observation of several nonlinear optical processes potentially useful for image processing and optical limiting applications; (ii) Fabrication of nonlinear liquid crystal filled 2- and 3-D photonic crystals and polymer dispersed liquid crystalline photonic crystals and demonstration of widely tunable transmission spectra; (iii) Investigation and characterization of the dynamics of the mechanisms responsible for the supranonlinearities of these liquid crystalline systems and identification of the roles of various fields, molecular and optical parameters for optimum nonlinearity, response time and other performance characteristics; (iv) Synthesis of neat nonlinear liquids with large two- and multi-photon absorption and excited-state absorption coefficients and characterizations of these liquids; (v) Demonstration of multiple time scale optical limiting capabilities of the organic liquids; (vi) Fabrication of nonlinear fiber arrays with these nonlinear liquids as wave-guiding core and evaluation of their image transmission capabilities; (vii) Quantitative modeling of self-action effects such as all-optical switching, optical limiting, nonlinear transmission of (fiber)guided visible and infrared lasers.

**Table 1 Electronic and Orientational Nonlinearities and Typical Response Times**

Electronic Nonlinearities	Nonlinear Coefficient [ $\beta \sim \text{Im } \chi^{(3)}$ ]	Response times
<b>Isotropic Liquid Crystals</b> [in neat liquid form]	Intrinsic value 5 cm/GW	$< 10^{-12}$ s
	Effective value $> 100$ cm/GW	$\sim 10^{-9}$ s – $10^{-12}$ s
* Damage Threshold: 10- 30 GW/cm <sup>2</sup> for visible & near IR nanosecond pulsed laser		
** Neat organic liquids are capable of ‘self-healing’ after damage, and can be easily infiltrated into micro-capillary arrays to form high quality image transmitting fiber array that could also function as large field of view optical limiter.		
Organic polymers [Most in solution form]	comparable or smaller	$< 10^{-12}$ s
Orientaional Nonlinearities	Index Coefficient n <sub>2</sub>	Response times [Typical]
<b>Nematic Liquid Crystals</b> [Very Broadband response]		
Supra-Nonlinear	$\sim 10^{-3}$ - 10	$\sim 10^{-3}$ s - $10^{-6}$ s
Photorefractive -nano doped	$\sim 10^{-3}$	$\sim 10^{-3}$ s
Order Parameter Change	$\sim 10^{-4}$	$\sim 10^{-6}$ s- $10^{-8}$ s
Photorefractive crystals and polymers	$\sim 10^{-3}$	$\sim 10^{-3}$ s
Semi-conductor photorefractive [narrow bandwidth]	$\sim 10^{-3}$	$\sim 10^{-6}$ s

The results and findings obtained in these projects are reported in 1 invention disclosure, 2 Ph. D. theses, 19 refereed journal papers and 21 technical conference presentations/proceedings, and in the new edition of a widely read/used reference book. This program has also resulted in active technology transfer and collaborative research with DOD laboratories and other government agency (e.g. DARPA), c.f. sections 5-8 of this report. In the remaining part of this section, we highlight the key breakthroughs obtained in this program of study and their significance.

## 2.2 Nonlinear nematic liquid crystals - Enhanced photorefractivity of CdSe nano-rod doped nematic liquid crystal

Photorefractivity in nematic liquid crystals was first discovered by this principal investigator [12]; subsequent work [9, 10] have demonstrated various aspects of this process including effective means for enhancing the effects and obtaining faster responses. A major accomplishment of this program is the demonstration of enhanced photorefractivity in nematic liquid crystals with the use of photosensitive nano-dopants such as metallic nano-wires and semiconductor nano-rods [11, 13]. Compared to other photorefractive inorganic and polymeric materials which require very large external applied fields [several 100's V/ $\mu$ m] to activate, nematic liquid crystals require much smaller applied field of <1V/ $\mu$ m, thus making them highly promising for practical device application.

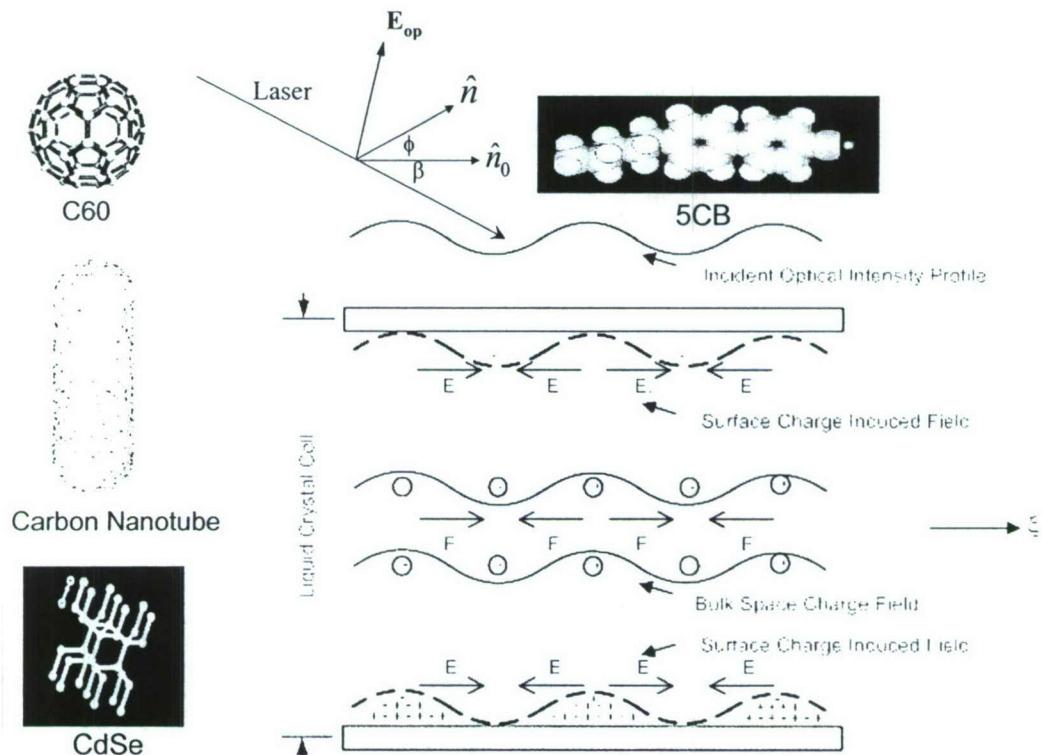


Fig. 1 Schematic depiction of the laser nematic interaction involved in a typical wave mixing process [e.g. photorefractive effect] in nano-particulates doped nematic liquid crystal (e.g. 5CB). Exemplary photosensitive dopants include CdSe nanorods, Fullerene C60 and Carbon Nanotube.

As schematically depicted in Fig. 1, an incident optical intensity distribution induces both bulk and surface space charge fields (1, 11). In liquid crystals the optically induced bulk space charge fields consist of 3 major components. One component,  $E_{ph}$  originates from photo-induced space charges and charge separation (diffusion and drift) and the other two,  $E_{\Delta\sigma}$  and  $E_{\Delta\epsilon}$ , are due to conductivity and dielectric anisotropies (Helfrich-Carr effect) and spatially reoriented director axis. Besides these bulk space charge fields, studies have demonstrated the important role played by the optically induced surface field modulation in lowering the threshold for initiating the photorefractive effect.

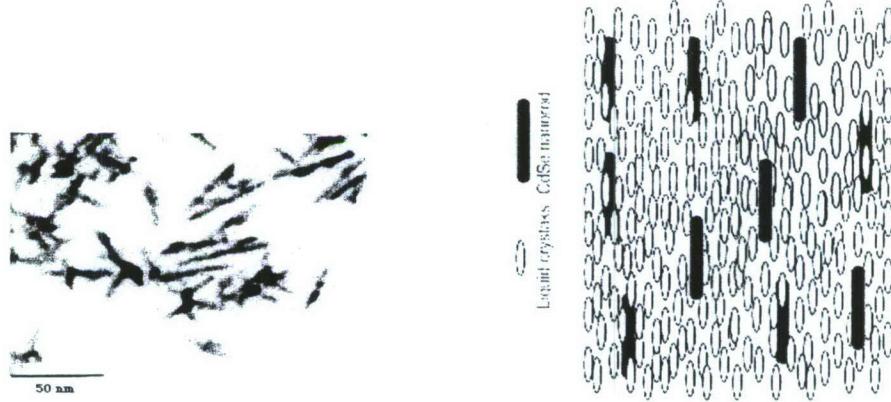


Fig.2 (left) SEM photo of CdSe nanorods. (right) Schematic depiction of planar aligned CdSe nano-rod doped nematic liquid crystal cell

Following our quantitative analysis [11] of the detailed torque balance equation governing the liquid crystal axis reorientation, in this case, the photorefractive threshold condition becomes:

$$\left[ E_{\Delta} E_{dc} \cos(\beta) + \left( \frac{\Delta\epsilon_{op}}{\Delta\epsilon} \right) E_{op}^2 \cos(2\beta) + E_s^{\text{eff}} E_{dc} \right] - E_F^2 \left[ 1 + \left( \frac{qd}{\pi} \right)^2 \right] > 0 \quad (1)$$

which leads to

$$E_{dc} > \left( \frac{\left( 1 + (qd/\pi)^2 \right)}{(\Delta\sigma/\sigma_{\perp} + \Delta\epsilon/\epsilon_{\perp}) \cdot \cos\beta} \right)^{\frac{1}{2}} E_F - \frac{1}{2(\Delta\sigma/\sigma_{\perp} + \Delta\epsilon/\epsilon_{\perp}) \cdot \cos\beta} E_s^{\text{eff}} \quad (2)$$

In these expressions,  $E_s^{\text{eff}} = \frac{E_s}{qd}$ ,  $\Delta\sigma$  is the conductivity anisotropy,  $\Delta\epsilon$  the dc dielectric anisotropy,  $\Delta\epsilon_{op}$  is the optical dielectric anisotropy,  $q = 2\pi/\Lambda$  is the grating wave vector with  $\Lambda$  the grating constant,  $E_s$  is the surface space charge field and  $d$  the cell thickness. In terms of the Freedericksz transition field  $E_F = \frac{1}{d} \sqrt{4\pi^3 k / \Delta\epsilon}$  [or voltage  $V_f = E_F d$ ], this gives:

$$V_{th} = \alpha V_F - \gamma E_s^{\text{eff}} d = \left( \alpha - \gamma \frac{E_s^{\text{eff}}}{E_F} \right) V_F \quad (3)$$

For 5CB,  $\Delta\epsilon \sim 11$  [ $\epsilon_{\parallel} \sim 16$ ,  $\epsilon_{\perp} \sim 5$ ],  $\Delta\sigma/\sigma_{\perp} \sim 0.5$ , and [ $qd \sim 2\pi$ , and the internal angle for a typical wave mixing geometry], equation (3) gives [11]:

$$V_F = 2.04V$$

$$V_{th} = \alpha V_F = 1.45 \times V_F \approx 3V \quad (4)$$

As remarked earlier, the required threshold voltages ( $\sim 3V$ ) for photorefractivity to be activated in nematic liquid crystals is much smaller and more practical than the  $\sim 1000$  V typically required in other inorganic or polymeric photorefractive crystals. Furthermore, by doping the nematics with charge-producing photosensitive dopants, e.g. carbon nanotubes, CdSe nano-rods, c.f. Fig. 2, the photorefractive effect in NLC can be dramatically enhanced, c.f. Fig. 3a.

Photorefractive effect in nematic liquid crystal requires that the optical electric field makes an inclination angle  $\beta$  with the initial director axis direction [1, 11]. To satisfy this requirement, one could employ planar aligned nematic cell [one in which the director axis is initially parallel to the cell wall] and apply an ac field above the Freedericksz transition to create a desired pre-tilt of the director axis [so that  $\beta$  is close to the optimal value of  $45^0$ ]. By adding a dc bias to the ac field, one can then generate the photorefractive effect with normal-incident writing laser beams, c.f. Fig. 3b. Not only does this configuration conform to practical image-processing requirements, it also provides a means of speeding the process since the ac field [and therefore the photorefractive effect] can be tuned-on and –off at a higher speed than free-relaxations

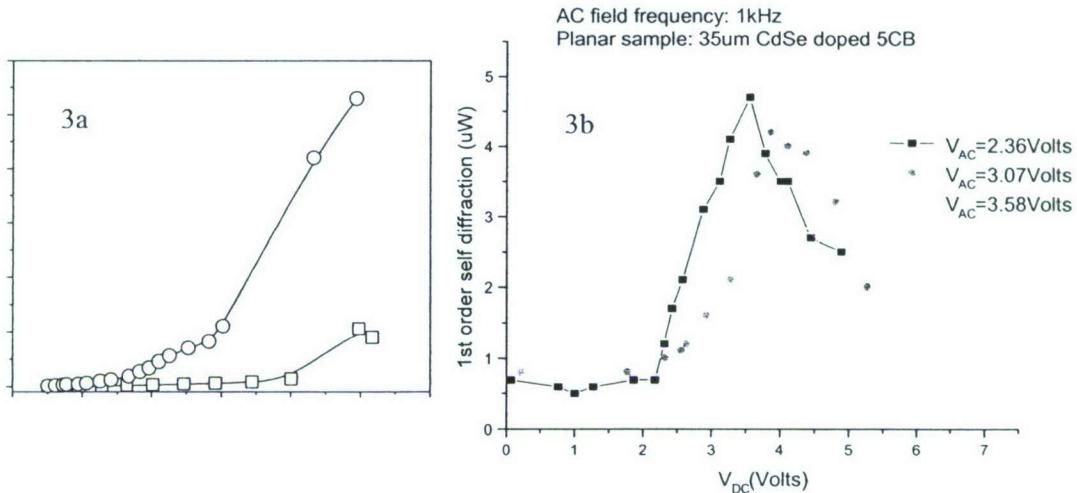
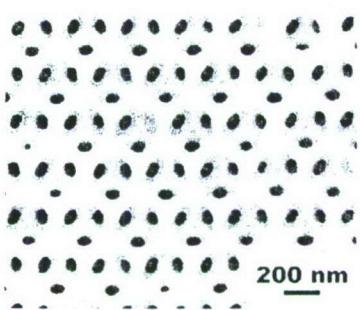


Fig.3 (a) An example of Enhanced four wave mixing efficiency [self-diffraction] of CdSe nano-rods doped liquid crystal (circles) over undoped one [squares]. (b) Self-diffraction signal from a planar aligned nematic liquid crystal under an applied ac field with various dc bias voltages.

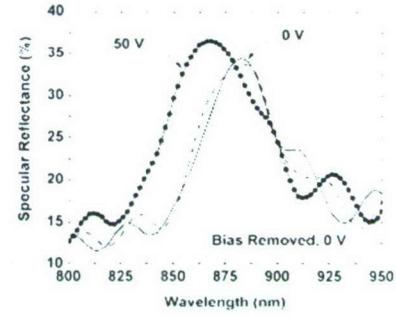
The observed nonlinear index coefficient  $n_2$  of CdSe-NLC is on the order of  $2.05 \times 10^{-2}$   $\text{cm}^2/\text{W}$ , which is more than 20 times larger than that of the undoped NLC. It is comparable to C60 and porphyrin:Zn doped NLC's and are orders of magnitude larger than all other nonlinear optical materials. These enhanced photo-refractive responses are

attributed to the increased charge generation capability of the photoconducting CdSe nanorods, as well as the larger dielectric and conductivity anisotropies  $\Delta\epsilon$  and  $\Delta\sigma$  of the doped nematic liquid crystals observed in recent experiments. It was also observed that under prolonged illumination, the induced orientation gratings remain transient in nature, unlike their C60 or dye (e.g. methyl red) counterparts which tend to produce unwanted persistent (or permanent) reorientation effect. This is attributed to the organic capping on the CdSe nanorods during the synthesis process; such capping inhibits adsorption of the optically excited CdSe nano-rods on the cell windows. This feature makes CdSe-doped NLCs promising candidates for real time image processing applications e.g. optical wave front conjugation, aberration correction and related adaptive optics applications.

Using these nonlinear optical responses of nano-modified liquid crystals, we have illustrated several all-optical and electro-optical switching operations in conjunction with visible – near infrared lasers. Exemplary work include: (i) Studies of nematic liquid crystals impregnated 3-D photonic crystals and demonstration of large [electrical] tuning range of their specular reflection [14], c.f. fig. 4a-b; (ii) Complete theoretical and experimental studies of passive all-optical switching and optical limiting with  $90^0$  twist alignment LC film [15]. The experimental observations are in good agreement with the theoretical expectations derived from a modified Jones Matrix calculation, and demonstrate the feasibility of designing efficient self-action optical limiting or laser hardening devices for long-pulsed and cw lasers; (iii) Tunable optical filters and reflective devices based on liquid-crystal-cladded frequency selective surfaces [16].



(a)



(b)

Fig. 4(a) SEM photo of a  $\text{TiO}_2$  inverse opal 3-D photonic crystal; (b) electrically tuning of Bragg reflection peak of the inverse opal structure containing aligned nematic liquid crystals.

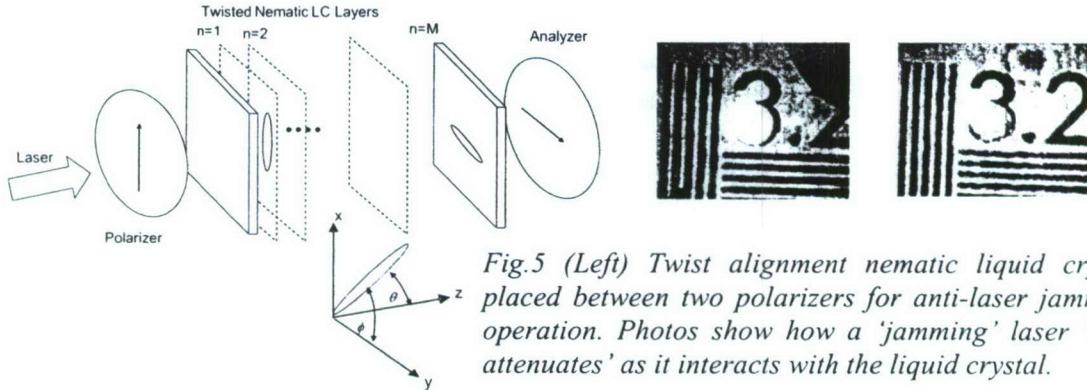


Fig.5 (Left) Twist alignment nematic liquid crystal placed between two polarizers for anti-laser jamming operation. Photos show how a 'jamming' laser 'self-attenuates' as it interacts with the liquid crystal.

Fig. 6a shows the computed transmission spectrum of a liquid-crystal-cladded FSS designed for the near infrared region [9]; for a liquid crystal birefringence  $\Delta n$  of 0.6, the planar FSS acts as a tunable filter with a tuning range of 380 nm. Since the birefringence of nematic liquid crystals span the **entire visible to far infrared spectrum**, c.f. Fig. 7, LC-FSS will allow us to design and fabricate very broadband high-extinction-ratio **tunable** filters and switches for polarized light by scaling up/down the FSS unit cells.

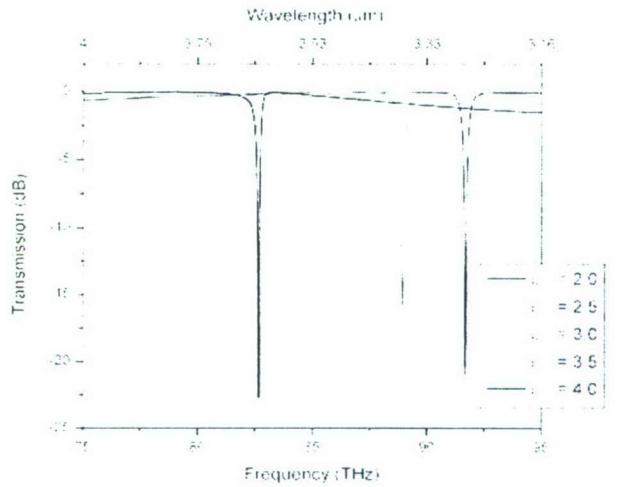


Fig.6 Transmission spectrum of a nematic liquid crystal (NLC) cladded-FSS structure for the range of dielectric constant of NLC showing very widely tunable optical filtering capability of the structure [ref.9]

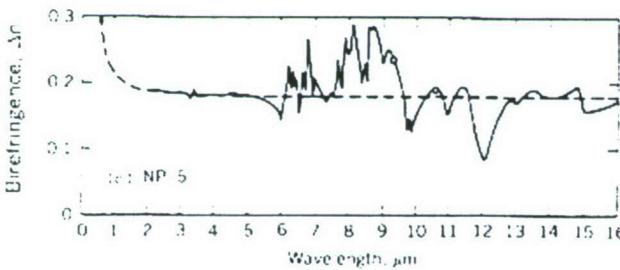


Fig.7 Typical birefringence of nematic liquid crystals [ref.1]

### 2.3. Extremely nonlinear multi-photon absorbing neat organic liquids and fiber arrays

In this program, we have synthesized several multifunctional nonlinear organic liquids that are optically transparent in the entire visible spectrum for low level light, but possess nonlinear multi-photon and excited-state-absorptions. Among them, the neat organic liquid L34 whose [linear] absorption spectrum and molecular structure are shown in Fig. 8a was found to possess multi-photon absorption properties that are ideally suited for optical limiting applications. Another class of nonlinear liquids that has been shown to be also effective for optical sensor protection against agile frequency lasers is ILC. ILC is a commercial product [from EM Chemicals, a subsidiary of BDH] that comprises four compounds [two of which are liquid crystalline at room temperature, whereas the other two are in the isotropic phase], c.f. fig. 8b. At room temperature, ILC behaves as isotropic liquid crystals, i.e. possess all the properties of a liquid. To enhance the nonlinear response of ILC, trace amounts of Fullerene C60 are dissolved in it to form C60-ILC. *An important recent breakthrough* in our laboratory is the identification/synthesis of the active component in ILC [which is no longer commercially available] and similar nonlinear liquids [derivatives of L34]. These liquids, called CE9'

and CE10', are non-volatile, freeze at much lower temperature [ $<-10^{\circ}\text{C}$ ] than L34 or ILC [L34 and ILC freeze at  $\sim 0^{\circ}\text{C}$ ], and are compatible with nano-particulate dopants such as C60 and CNT for enhanced performance. From previous nanoseconds and recent femtoseconds-picoseconds studies, L34 possess a molecular photonic level structure uniquely suited for optical limiting application. As depicted in Fig.9, starting from the ground state, the first non-vanishing transition is via a two-photon absorption process to level 2. From there the molecule could undergo either a direct single photon transition to high lying excited state, and/or intersystem crossing to a triplet manifold followed by a transition to higher excited states [17, 18].

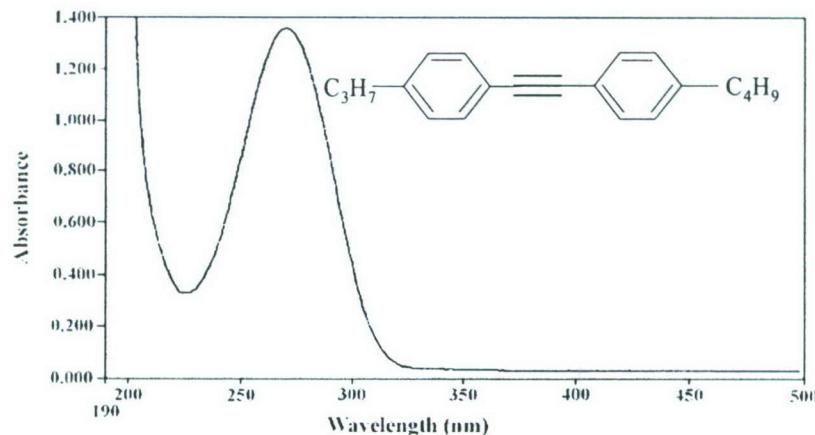


Fig.8a Molecular structure and absorption spectrum of L34

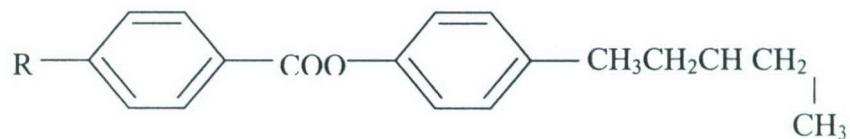


Fig.8b. Molecular structure of ILC - a mixture of 4 organic liquids [where R designates:  $\text{C}_6\text{H}_{13}$ ,  $\text{C}_8\text{H}_{17}$ ,  $\text{C}_3\text{H}_7$ ,  $\text{C}_5\text{H}_{11}$ ]. It has similar absorption spectrum as L34.

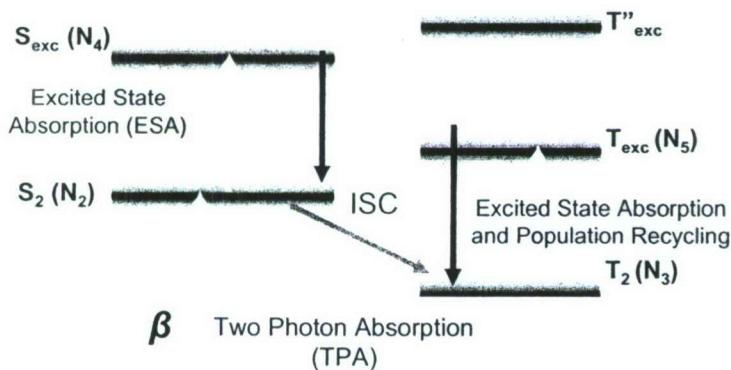


Fig. 9 Schematic depiction of the molecular energy level structure of LC4 and various single-,two-photon absorption (TPA) and excited state absorption (ESA).

The characteristic time constants for intersystem crossing are in the nanosecond regime. As a result, nanosecond nonlinear transmission measurements yield effective absorption coefficients much larger than their intrinsic values. For example, the intrinsic TPA coefficient for L34 measured with picosecond laser pulses [see next section] is  $\beta \sim 0.75 \text{ cm/GW}$ . On the other hand, measurements using nanosecond laser pulses, yield an intensity dependent effective two photon absorption coefficient  $\beta_{\text{eff}}$  ranging from 15 cm/GW at low laser energy [2.5  $\mu\text{J}$ ] to more than 170 cm/GW at high laser energy [about 30  $\mu\text{J}$ ], c.f. Fig. 10. An estimate of the excited state absorption coefficient  $\alpha_{\text{exc}}$  can be made from these measurements using a theoretical formalism we have developed. For L34,  $\alpha_{\text{exc}} \sim 1.7 \times 10^4 \text{ cm}^{-1}$ . As reported in [17, 18], these extraordinarily large effective nonlinear absorption coefficients are responsible for the efficient optical limiting performance of L34 against nanosecond laser pulses.

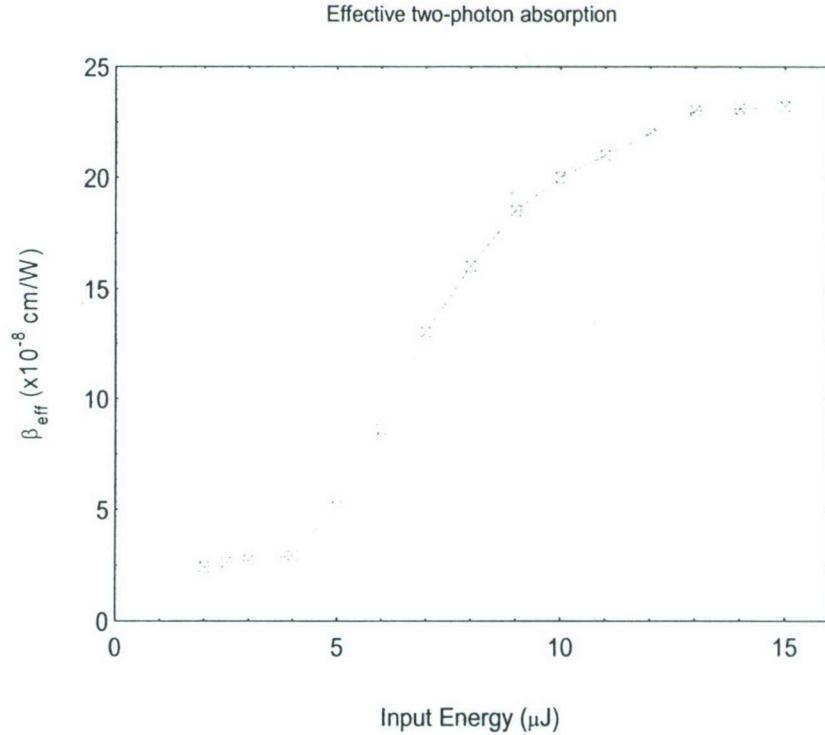


Fig. 10a: Observed intensity dependent effective two-photon absorption coefficient  $\beta_{\text{eff}}$ . Laser pulse width: 7ns; wavelength: 532 nm; focused spot radius  $\omega_0 = 6 \mu\text{m}$ ; bulk liquid film thickness: 0.1 mm.  $\beta_{\text{eff}}$  is much larger than the intrinsic value  $\beta_{\text{eff}}$  and is attributed to ESA.

In order to measure the intrinsic two-photon absorption coefficient of L34 more accurately, we employ picosecond and femtosecond laser pulses. In this time scale that is shorter than the intersystem [singlet  $\rightarrow$  triplet] crossing, direct [singlet manifold] excited state absorption could still come into play. This is indeed observed in some very recent nonlinear transmission experiment with L34 where the (direct singlet state) excited state contribution following two-photon absorptions from the ground state give rise to an intensity dependent effective absorption coefficient [18]. Fig. 10b depicts the intensity dependent (increasing) absorption effects obtained from a standard z-scan transmission set up for a 0.5 mm thick bulk L34 cell and Fig. 10c shows the fitted nonlinear absorption

coefficient's intensity dependence. This yields  $\beta_{\text{eff}}$  varying from 0.75 cm/GW at a low input intensity to  $\sim 4.25$  cm/GW at an input intensity of 33 GW/cm<sup>2</sup>. The intercept at vanishing value of the laser intensity provides an estimate of the intrinsic two-photon absorption coefficient  $\beta \sim 0.75$  cm/GW. The presence of direct single-photon excited state absorption to the two-photon absorption from the ground state will certainly contribute to the limiting performance of L34 against picosecond laser pulses and is being incorporated in ongoing theoretical modeling efforts.

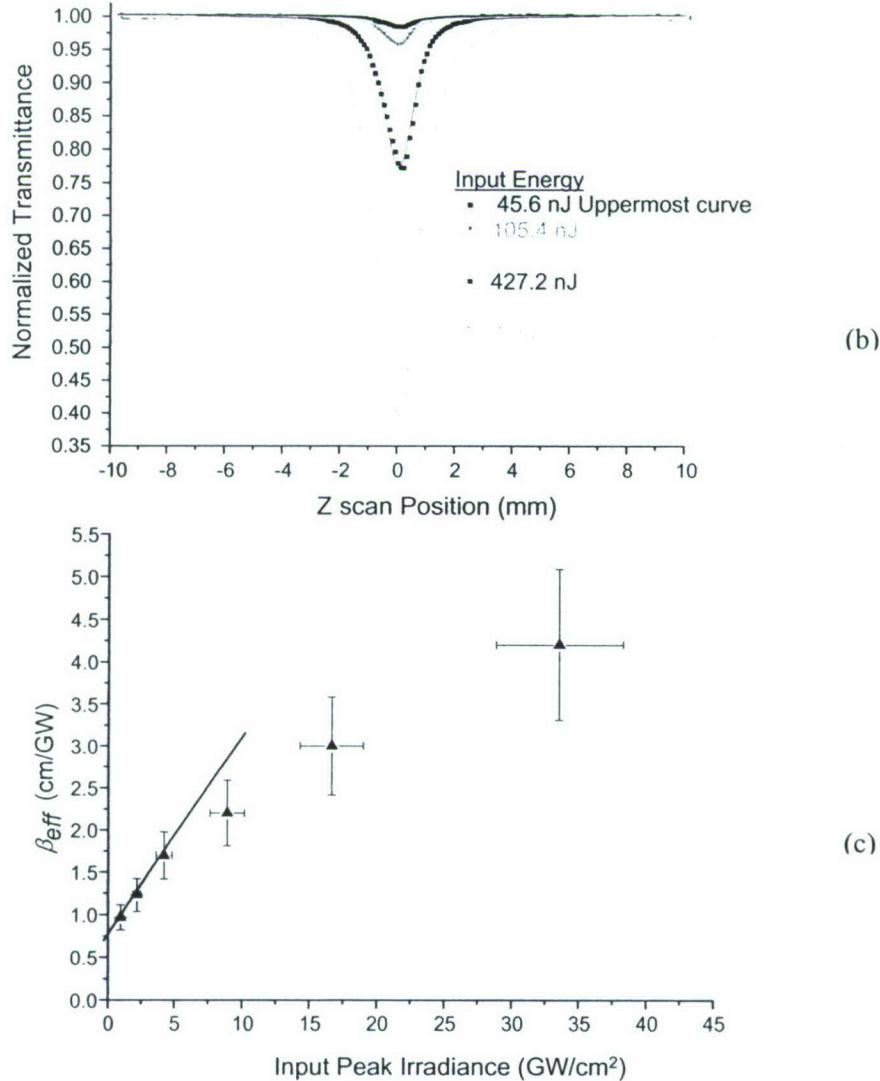
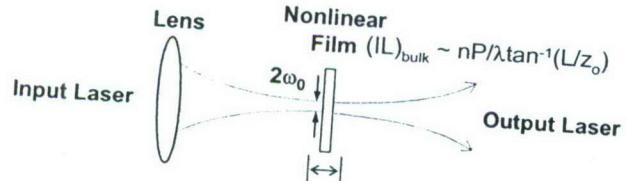


Fig. 10(b) Nonlinear transmission of L34 obtained by Z-scan technique; (10c) Effective nonlinear absorption coefficient of L34 as a function of the input laser peak intensity of the laser. Experiment conducted at CREOL, UCF with picosecond laser. Experimental parameters: 14 ps FWHM,  $\omega_0 = 10.5 \mu\text{m}$ ; wavelength = 532 nm. Sample thickness: 0.5 mm.

Another major success in our program is the feasibility demonstration of a fiber array made with these nonlinear liquids as the waveguiding cores. The fiber arrays with nonlinear absorbing cores possess many advantageous features over conventional 'free-space' optics. In nonlinear absorbers such as TPA materials, qualitatively speaking, the

transmitted laser intensity  $I_{out}$  is related to the input intensity  $I_n$  by  $I_{out} \sim I_n \exp -\beta(IL)$ , where  $(IL)$  is the integrated value of the laser over the interaction length  $L$  and  $\beta$  is the two-photon absorption constant, for example. For a given incident laser power  $P$ , the  $(IL)$  value for a 'free-space' focusing optics in a bulk nonlinear liquid is given by:  $(IL)_{bulk} \sim nP/\lambda \tan^{-1}(L/z_0)$  where  $z_0 = \pi n \omega_0^2/\lambda$ , c.f. Fig. 11. On the other hand, in fiber, we have:  $(IL)_{fiber} \sim PL/\pi a^2$ . With the confined/guided-wave propagation geometry, the fiber array will provide a much larger  $(IL)$  value. For example, for  $L \sim 5$  mm,  $\lambda = 0.532 \mu\text{m}$ ,  $a = \omega_0 \sim 15 \mu\text{m}$ ,  $n=1.5$ , we have  $(IL)_{fiber}/(IL)_{bulk} \sim 7$ . In F1 optics used in many sensor systems, the focused beam radius can be smaller (e.g.  $\omega_0 \sim 5 \mu\text{m}$ ), and the corresponding fiber advantage factor will be much larger. This translates to lower limiting threshold as well as clamped output in fiber versus bulk materials, and has been demonstrated with L34 [17, 18].



For  $\omega_0=5 \mu\text{m}$ ,  $\lambda=532 \text{ nm}$ ,  $L_{int}=0.3 \text{ mm}$

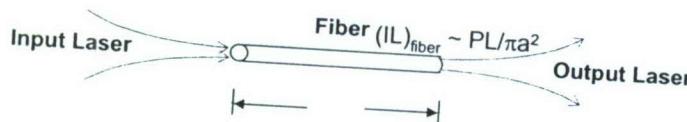


Fig. 11 Illustration of the extended nonlinear interaction length in fiber geometry versus free-space focused optics in bulk film

In the course of our studies, we have fabricated a variety of fiber arrays by filling glass capillaries with L34. The refractive index of L34 is 1.62 and the glass capillary is 1.51. The fiber (capillary) core diameter ranges from 20 - 30  $\mu\text{m}$  and the lengths are in the range of 2 - 4 mm. When placed in the focal plane of a 1x telescope, c.f. Fig. 12, the

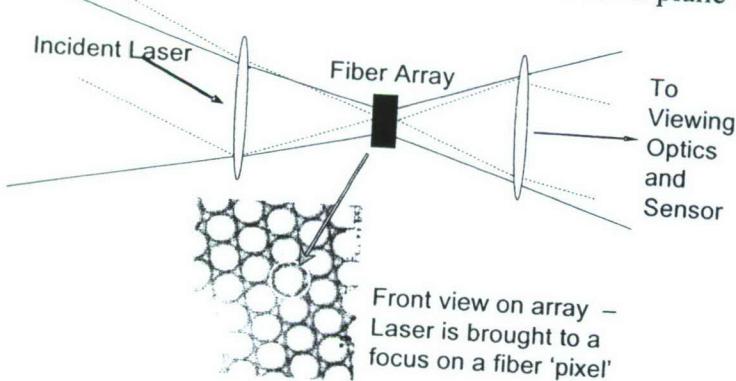


Fig. 12 Imaging and optical limiting actions of a fiber array placed in a 1x telescopic optical system. Photograph shows a WPAFB courtyard imaged through the system.

scenery is imaged onto an area covering many fibers, whereas the laser, originating as a point source and impinges on the fiber array from a particular direction (spatial

frequency), will be imaged (focused) onto a single fiber [a pixel on the imaging plane] by the front end fast optics (usually small F#’s lens). Because of the large difference between the core and cladding refractive index, i.e. large numerical aperture, the field of view (FOV) of these L34-cored fiber arrays can be very large. For example, for a core diameter of 20  $\mu\text{m}$  and refractive indices  $n_{\text{core}}=1.62$  [for L34] and  $n_{\text{cladding}}=1.51$  (for the glass capillary), the field of view is  $\sim 40$  degree. Better image resolution can be achieved with smaller fiber core diameters, similar to conventional fiber faceplates.

When used as a sensor protection device, nonlinear mechanisms such as two-photon and excited state absorptions by the fiber core liquid will act in concert to clamp the transmission of an incident (high intensity) laser, while letting the low light level scenery to pass un-attenuated. *This intensity dependent spatial frequency filtering ability of the fiber array, coupled to the longer interaction length and much better light blocking action of the opaque cladding, result in excellent optical limiting performance impossible with bulk material and free-space optics.* In the (fiber) geometry, other limiting mechanisms such as self-defocusing, nonlinear scattering and guided mode loss due to index change at high laser intensity also contribute to limiting the transmission of the incident laser, c.f. Fig. 13a. This and other advantages unique to fiber geometry enable low switching thresholds, large dynamic range and other limiting performance better than other material/device geometry.

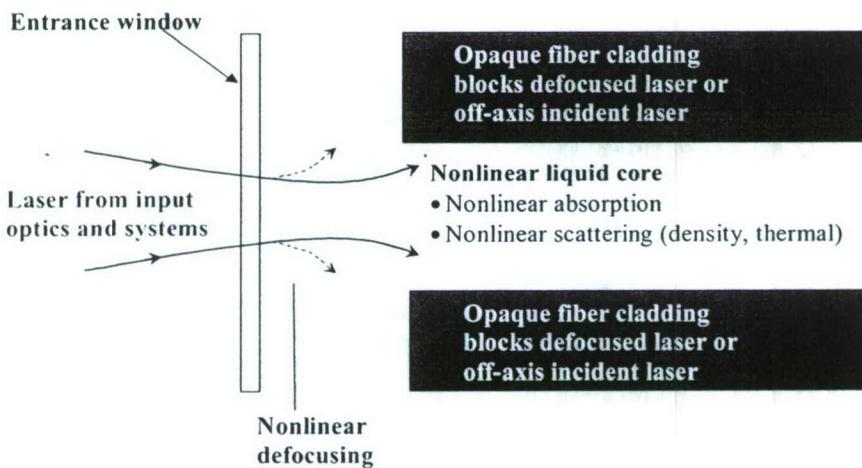


Fig. 13a Nonlinear optical processes responsible for optical limiting actions of L34 cored fiber array.

Fig.13b shows exemplary optical limiting results obtained recently with nanosecond pulsed laser [wavelength = 537 nm] through a 4-mm L34 cored fiber array [core diameter = 30  $\mu\text{m}$ ]. The laser intensity at the output end of the fiber is monitored [data in circles]. The date show that for laser energies as high as 0.3 mJ (corresponding to an input fluence of  $\sim 30 \text{ J/cm}^2$  for the fiber core diameter of 30  $\mu\text{m}$ ), the transmitted laser fluence arriving at the sensor at the output end of the fiber is clamped to  $< 0.2 \text{ J/cm}^2$  - which is much lower than typical sensors’ Maximum Permissible Exposure (MPE) value of  $3 \text{ J/cm}^2$  for nanosecond laser pulses. It is important to point out that the low-laser-energy transmission of the transparent L34 in the fiber core is over 70 %, with the loss mainly coming from input coupling loss and reflections from the uncoated optics used.

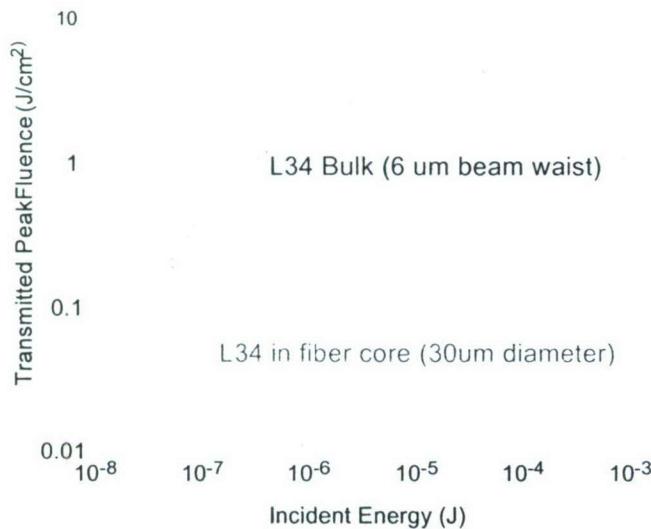


Fig.13b Optical limiting results with 4-mm thick bulk L34 and L34-cored fiber array obtained at WPAFB.

It is interesting to note that because of the large effective nonlinear absorption coefficient of L34 in the nanosecond time scales, even bulk liquid [4-mm thick] could clamp the transmission of the nanosecond laser pulses below the MPE value of sensors. For the case of a laser focused into the back plane of a 4-mm thick bulk L34, an input fluence of  $30 \text{ J/cm}^2$  (corresponding to an input laser energy of  $1.1 \mu\text{J}$  for a laser spot diameter of  $6 \mu\text{m}$ ), gives a clamped output fluence of  $\sim 2 \text{ J/cm}^2$ , which is  $< \text{MPE}$  [see Fig.13 data plotted in squares]. Bulk L34's are therefore promising candidates for applications in optical systems where there is no need for fiber imaging faceplate.

#### 2.4 Longer Time Scale [microseconds – CW] Optical Limiting Actions

Although nonlinear absorbers of the type discussed in the preceding section [RSA, TPA, TPA+RSA] are highly desirable in many ways, there is nevertheless an inherent limitation. Basically, these nonlinear absorbers exploit the higher intensity [for a given laser pulse energy] of short [nanosecond or sub-nanosecond] laser pulses to initiate optical self-action effects. Clearly, the efficiencies of these processes will rapidly deteriorate as the laser pulse intensity is reduced as a result of longer pulse durations. The problem becomes more acute in situations involving sensor jamming or 'white-out' caused by cw lasers or glares. Accordingly, in the 10's of nanosecond and longer time scale, other nonlinear mechanisms such as thermal and density effects have to be invoked. Fortunately, organic neat liquids such as L34, CE9' and CE10' [in undoped or doped forms] exhibit large thermal/density effects [through intra- and/or inter-molecular non-radiative relaxations] and the characteristic time constants for these effects to manifest are precisely in this time scale of concern. In this regards, the nearly complete transparency of these neat liquids developed in our program is particularly important as it allows one to introduce a variety of highly appropriate dopants to enhance these nonlinear responses without incurring severe transmission loss.

To account for the presence of thermal and density effects, we have initiated the development of complete nonlinear pulse propagation model by including the laser induced temperature and density fluctuations ( $\Delta T(\mathbf{r},t), \Delta \rho(\mathbf{r},t)$ ) in the nonlinear fiber propagation model that accounted for molecular nonlinear absorption process. Besides

the molecular energy level populations  $N$ 's, we have to consider additionally the coupled hydrodynamic equations for  $\Delta T(\mathbf{r}, t)$  and  $\Delta \rho(\mathbf{r}, t)$ :

$$-\frac{\partial^2}{\partial t^2}(\Delta \rho) + v^2 \nabla^2(\Delta \rho) + v^2 \beta_T \rho_0 \nabla^2(\Delta T) + \frac{\eta}{\rho_0} \frac{\partial}{\partial t} \nabla^2(\Delta \rho) = \frac{\gamma^e}{8\pi} \nabla^2(E^2) \quad (6)$$

and

$$\rho_0 C_v \frac{\partial}{\partial t}(\Delta T) - \lambda_T \nabla^2(\Delta T) - \frac{(C_p - C_v)}{\beta_T} \frac{\partial}{\partial t}(\Delta \rho) = \frac{u}{\tau} = \frac{\alpha n c}{4\pi} E^2 \quad (7)$$

where  $\rho_0$  is the unperturbed density of the liquid crystal,  $C_p$  and  $C_v$  the specific heats,  $\lambda_T$  the thermal conductivity,  $\eta$  the viscosity,  $v$  the speed of sound,  $\gamma^e$  the electrostrictive coefficient  $[\gamma^e = \rho_0 (\rho \varepsilon / \partial \rho)_T]$ ,  $\beta_T$  the coefficient of volume expansion, and  $\eta$  a viscosity coefficient.

Some preliminary experiments with these isotropic liquids doped with fullerene C60 [a RSA absorber] show that indeed they are highly promising materials of choice for sensor protection or optical limiting application against microsecond and cw lasers, c.f. Fig. 14. These results also illustrate how one could extend the operation bandwidth from the visible to the near IR (750 nm) and communication wavelength (1550 nm) regime with the use of readily available nano-dopants such as fullerene C60.

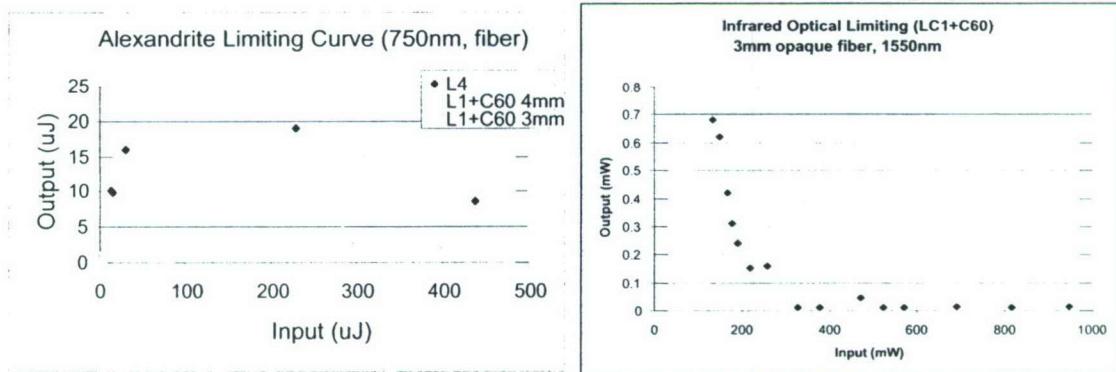


Fig. 14 Preliminary optical limiting studies of fiber cores containing pure and doped isotropic liquid crystals with 750 nm microsecond pulsed laser and 1550 nm cw laser.

Recently, we have demonstrated successful fabrication of Au and Ag nanospheres that could be dispersed in L34 to achieve similar optical limiting performance with visible as well as near IR lasers in the nanoseconds – microseconds time scales [19]. Fig. 15 shows a plot of the output vs. input of q-switched (300 ns full width) 750 nm laser through a 2 mm thick Au-L34 sample in a closed aperture set-up. The concentration of Au is chosen such that the linear transmission of the sample is  $\sim 70\%$ . The laser is focused by a lens of focal length 5 cm to an estimated spot size of  $\sim 100 \mu\text{m}$ . The size of an aperture placed downstream in a F6 set-up allows the low energy ( $< 1 \mu\text{J}$ ) laser pulses to pass through. At high power, it is observed that the laser is strongly scattered and defocused. The 2 mm bulk cell, with the help of the aperture in front of the downstream detector, is able to clamp the transmission to  $\sim 2 \mu\text{J}$  for input up to 2 mJ.

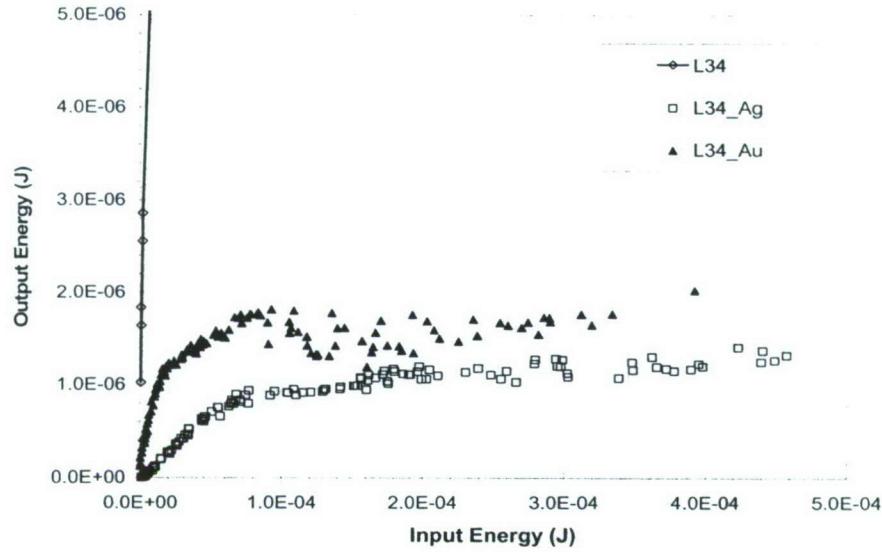


Fig. 15 Plot of the output versus input of a 750 nm Q-switched lasers (~100 ns duration) through a 2-mm thick Au (diameter ~ 2.5 nm) and Ag (diameter ~ 4-5 nm) nano-sphere doped L34 bulk liquids. Almost linear curve is from pure L34.

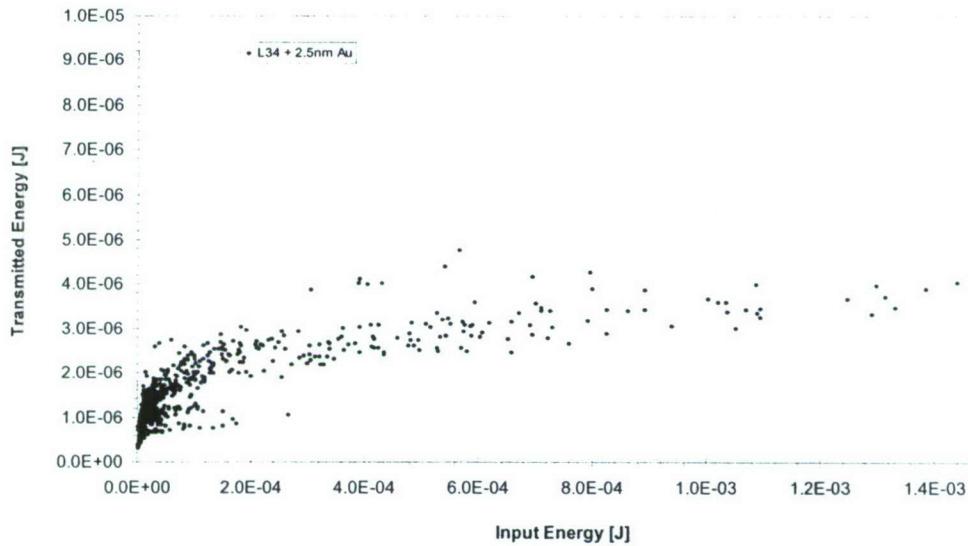


Fig. 16 Plot of the output versus input of a 750 nm free running (30  $\mu$ s) pulsed lasers through a 2-mm thick Au (diameter ~ 2.5 nm) nano-sphere doped L34 bulk liquid.

We have repeated the experiment with the laser operating in the (multiple-spikes) free-running mode (30  $\mu$ s total pulse duration), and observed similar nonlinear transmission performance, c.f. Fig. 16. Au-doped L34, therefore, could be used for clamping pulsed lasers over the vast time scale ranging from picoseconds through nanoseconds to microseconds. Isotropic nonlinear neat liquids, in conclusion, are highly promising candidate in our quest to develop single fiber array optical limiter that can be effective over a very large time-scale, and will therefore be the major thrust in our proposed new program.

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\*20. I. C. Khoo, "Supra-nonlinear nano-dispersed liquid crystalline negative and zero-index optical meta-materials," Invited paper, 12<sup>th</sup> Int. Topical Meeting on Optics of Liquid Crystals," Puebla, Mexico, Oct. 1-5, 2007.

\*21. I. C. Khoo and A. Diaz, " Liquid Crystal Optical Metamaterials, " IEEE -LEOS Winter Topical 2008, Sorrento, Italy, Jan. 2008.

## 7. Inventions Disclosures and Book:

- [Invention disclosure] I. C. Khoo and A. Lei "Extremely low freezing point non-volatile nonlinear optical liquids for eye/sensor protection against intense laser pulses" PSU Invention Disclosure #2002-2707 #2003-2854

- [Book] I. C. Khoo, Liquid Crystals, 2<sup>nd</sup> Edition (Wiley, NJ 2007)

## 8. Interactions with DOD Laboratories and Development Centers

(i) Wright Patterson Air Force Base [Tim Bunning, Paul Fleitz, Joy Rogers and Augustine Urbus]:

We have engaged in active technical discussions and collaborative research activities on various aspects of this program. These collaborations are centered on using nonlinear liquid crystalline films for optical filters, switches and laser hardening applications. Recent collaborations are centered on femtosecond and picosecond characterization of highly nonlinear organic neat liquids that possess Two-Photon Absorption [TPA] and Excited State Absorption [ESA]. Using various pump-probe and single- and two-photon induced fluorescence and nonlinear transmission measurements, Paul fleitz and Joy Rogers have measured the excited state absorption spectrum and two-photon induced fluorescence/absorption spectra of L34. The objectives are to evaluate the performance limits of these organic neat liquids as fiber core for optical limiting applications against short [nanosecond] laser pulses. Recently, Augustine Urbus has started a series of optical limiting tests with L34 bulk and fiber arrays, as well as Au-L34 for optical limiting of nanoseconds – microseconds lasers.

(ii) Navy Air Development Center, Patuxent River, MD [Jim Sheehy]; Defense Advanced research Projects Agency [Dave Shenoy]:

We had a cooperative agreement to develop Helmet-mount eye/sensor laser protection goggle for Navy pilots in collaboration with the SRI International, Menlo Park, CA] against agile frequency lasers throughout the visible regime. The project was completed 7/2006, and the device development efforts have been transitioned to a DARPA phase II and Phase III projects on near IR sensor protection device development.

(iii) - Consultant to a Army Research Office SBIR Phase II program on nano-dispersed supra-nonlinear liquid crystals for spatial light modulation awarded to BEAM Engineering Corporation, Winter Park, Florida. Completed 8/2005.

- Consultant to a Air Force Research Laboratory [WPAFB] SBIR Phase II program on sensor protection and optical limiting awarded to BEAM Engineering Corporation, Winter Park, Florida. Project completed in 9/2005.

## 9. Graduate Theses

### **Ph. D. Theses:**

1. Yana Z. Williams (2006)

ThesisTitle: Electro- and nonlinear- optics of liquid crystals with nano-dopants and nano-structures

2. Kan Chen (2006)

Thesis Title: Photorefractive nonlinearity in pure and doped liquid crystals

### **MS thesis:**

Mike Stinger (2006)

Thesis Title: Stimulated Orientational Scattering